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Biogeosciences Discussions, 2, 1–35, 2005
www.biogeosciences.net/bgd/2/1/
SRef-ID: 1810-6285/bgd/2005-2-1
European Geosciences Union

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Received: 27 December 2004 – Accepted: 6 January 2005 – Published: 10 January 2005

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Abstract

Submarine groundwater discharge was quantified by a variety of methods in Salt Pond, adjacent to Nauset Marsh on Cape Cod, USA. Discharge estimates based on radon and salinity took advantage of the presence of the narrow channel connecting Salt Pond to Nauset Marsh, which allowed constructing whole-pond mass balances as water flowed in and out due to tidal fluctuations. A box model was used to estimate discharge separately to Salt Pond and to the channel by simulating the timing and magnitude of variations in the radon and salinity data in the channel. Discharge to the pond is estimated to be $2200\pm1100\,m^3\,d^{-1}$, while discharge to the channel is estimated to be $300\pm150\,m^3\,d^{-1}$, for a total discharge of $2500\pm1250\,m^3\,d^{-1}$ to the Salt Pond system. This translates to an average groundwater flow velocity of $3\pm1.5\,cm\,d^{-1}$. Seepage meter flow estimates are broadly consistent with this figure, provided discharge is confined to shallow sediments (water depth <1 m). The radon data can be modeled assuming all groundwater fluxes to both the channel and to the pond are fresh, with no need to invoke a saline component. The absence of a saline component in the radon flux may be due to removal of radon from saline groundwater by recent advection of seawater or it may to due to the presence of impermeable sediments in the center of the pond that limit seawater recirculation. This groundwater flux estimated from the radon and salinity data is comparable to a value of 3200–4500 $m^3\,d^{-1}$ predicted by a recent hydrologic model (Masterson, 2004; Colman and Masterson, 2004[1]). Additional work is needed to determine if the measured rate of discharge is representative of the long-term average, and to determine the rate of groundwater discharge seaward of Salt Pond. Data also suggest a TDN flux from groundwater to Salt Pond of $\sim2.6\,mmol\,m^{-2}\,d^{-1}$, a figure comparable to fluxes observed in other eutrophic settings.

[1]Colman, J. A. and Masterson, J. P.: Transient nutrient load simulations for a coastal aquifer and embayment, Cape Cod, Massachusetts, Environ. Sci. Technol., submitted, 2004.

1. Introduction

In recent years it has become increasingly clear that submarine groundwater discharge (SGD) has a significant impact on the coastal ocean (Moore, 1996; Burnett et al., 2003). Some of the earliest research on SGD sought to quantify its role in the delivery of nutrients to the coastal ocean (e.g. Valiela et al., 1990; Giblin and Gaines, 1990), and this continues to be an important focus, particularly in places where domestic wastewater is treated by septic systems. However, because the influence of SGD on the ocean has only recently been recognized, there has also been a need to study the processes (e.g. redox, microbial, mixing of fresh and saline waters) that transform elements in the "subterranean estuary" (*sensu* Moore, 1999) (e.g. Charette and Sholkovitz, 2002). In addition, there has been a need for basic research aimed at improving our understanding of the contribution of SGD to the marine budget of certain elements (e.g. Fe, Ba, Ra).

It is important that we define what we mean by "submarine groundwater discharge" at the onset of this paper. We will use the term SGD, as defined by Burnett et al. (2003), to refer to "any and all flow of water on continental margins from the seabed to the coastal ocean, regardless of fluid composition or driving force." This definition includes both fresh groundwater and circulation of seawater through sediments, and is thus not equivalent to the traditional concept of fresh groundwater as defined by terrestrial hydrologists. Later in this paper we will address what various tracers can tell us about the fresh and saline components of SGD.

Quantifying SGD remains difficult, despite our increased awareness of its importance, because discharge is diffuse and heterogeneous and occurs below the water surface, where direct observation and measurement are difficult. Nonetheless, three primary methods have arisen in recent attempts to quantify SGD: 1) groundwater flow models; 2) seepage meters and 3) radioisotopes (radon and radium). Salinity can also serve as a tracer of fresh groundwater discharge in settings where groundwater represents the only source of fresh water.

In some locations, groundwater flow models are sufficiently well developed that they can be used to predict the delivery of fresh groundwater to the coast. While in these settings the hydrologic budget may be well constrained such that total discharge over a large coastal area is well known, the precise location of discharge of that freshwater in the coastal zone is often not known. Furthermore, it is worth noting that one recent study from the Northeast Gulf of Mexico (Smith et al., 2003) concluded that model-based fresh groundwater discharge estimates were much lower than field-based estimates based on radioisotopes and seepage meters unless the hydraulic conductivity was much higher than considered in the model. Hence, there is a need for more comparisons between modeled and measured discharge estimates, and a need for additional methods to measure the locations, and the rates of discharge.

Seepage meters have been used to quantify groundwater discharge below the water surface for many years (Lee, 1977). However, at their best, seepage meters only yield an average discharge rate spanning the small area of deployment (typically $<1\,m^2$). Because discharge is often heterogeneous, many seepage meters are needed to yield discharge estimates representative of a large area.

Yet another approach for quantifying the flux of SGD involves the use of radioisotopes, specifically radium and radon. Each of these elements has been used as a tracer of groundwater discharge in the coastal zone because each is enriched in groundwater, relative to surface water, often by two to three orders of magnitude (Burnett and Dulaiova, 2003). Both radium (Ra) and radon (Rn) are members of the ^{238}U decay series. Spanning the most recent tens to hundreds of thousands of years, the lineage of radon and radium is as follows: ^{230}Th ($t_{1/2}$=75 000 years) decays to ^{226}Ra ($t_{1/2}$=1600 years) which in turn decays to ^{222}Rn ($t_{1/2}$=3.8 d). In this work we focus on the use of radon, primarily because radon behaves conservatively spanning the salinity range from freshwater to seawater. This simplifies coastal zone interpretations where large salinity gradients are common. By quantifying the flux of radon to coastal waters and the radon content of the local groundwater, an estimate of SGD can be derived, as will be elaborated later in this paper. One strength of this approach is that measurements of the radon flux to surface waters integrate over a large area; hence, SGD estimates inferred in this way integrate over the same large area.

It is worth noting that the use of radon as a tracer of groundwater discharge to surface waters is not new, dating to work by Ellins et al. (1990). However, the field has been advanced in recent years by new technologies permitting continuous radon measurements using the RAD7 radon analyzer (Burnett et al., 2001; Burnett and Dulaiova, 2003).

This paper describes an intercomparison of several methods of quantifying SGD, carried out in Salt Pond, a saline, drowned kettle hole pond at the northern end of Nauset Marsh (Fig. 1). The primary focus of this work is estimating SGD to Salt Pond using continuous measurements of radon and salinity, together with a box model. We will also describe measurements made with seepage meters. Each of these approaches yields an estimate of SGD that can be compared to predictions derived from a mature groundwater flow model that has been written for the eastern portion of Cape Cod (Colman and Masterson, 2004; Masterson, 2004). In addition, nutrient fluxes from groundwater to Salt Pond are estimated and compared with fluxes from other locations.

2. Sampling locations and methods

Salt Pond is a saline, drowned kettle hole pond at the northern end of Nauset Marsh within Cape Cod National Seashore, USA (Fig. 1). Salt Pond is roughly circular, with a surface area of 82 200 m^2, a maximum depth of roughly nine meters and a mean depth of 3.4 m (Anderson and Stolzenbach, 1985). There is no surface runoff to the pond, which is connected to Nauset Marsh by a channel that is roughly 30 m wide at low tide, 350 m long, and 0.6 m deep at the thalweg (low point) at low tide.

The following brief summary of the hydrogeologic setting of Salt Pond is derived from Masterson (2004) and Colman and Masterson (2004), to which we refer the reader for greater detail. Groundwater flow to Salt Pond is derived from the Lower Cape Cod aquifer, which contains sediments deposited at the end of the last glacial period that range in size from clay to boulders. The hydraulic conductivities range from $0.0035\,\mathrm{cm\,s^{-1}}$ in clay to $0.07\,\mathrm{cm\,s^{-1}}$ in gravel, with the ratio of horizontal to vertical conductivity ranging from 5:1 in coarse material to 100:1 in fine-grained material.

Between 28 June and 2 July 2004, radon, salinity, temperature and water depth were measured within the channel between Salt Pond and Nauset Marsh. Radon measurements were carried out from a raft that was anchored at the northern end of the channel (Fig. 1) using methods similar to those described in Burnett et al. (2001). Briefly, the method involves pumping water at a flow rate of $\sim 2\,\mathrm{L\,m^{-1}}$, stripping the radon into the gas phase, and measuring ^{218}Po, a decay product of radon, using a RAD7 radon detector (Burnett et al., 2001).

The pump failed on a few occasions when an inline strainer (0.5 mm pore size) clogged due to the presence of significant algal biomass. As a result, no radon data were obtained during these intervals when there was no water circulation. However, the precise times when the pump failed could be determined after the experiment as times when the temperature measurements of the water pumped for radon measurements (thermistor exposed to air during pump failure) differed from temperature measurements from the CTD (in the channel) by more than a few tenths of a degree.

A weather station (Onset HOBO), also attached to the raft, recorded wind speed at 5 min intervals using a propeller-type anemometer mounted at a height of 2.3 m above the water surface. Wind speeds were converted to a height of 10 m following the method of Donelan (1990), assuming a neutrally stable boundary layer, a logarithmic wind profile and a drag coefficient at 10 m height of 1.3×10^{-1} (Large and Pond, 1981).

Salinity (S) and temperature (T) were measured in a variety of locations and times. The salinity, temperature and depth of the water in the channel were measured and logged every 5 min using a YSI 600XLM Sonde positioned twenty three cm above the channel bottom in water $\sim$0.6 m deep at low tide, roughly eight m towards Nauset Marsh from the raft position. The calibration of the YSI salinity data was carried out using discrete samples analyzed at the Woods Hole Oceanographic Institution (WHOI) CTD calibration facility using a Guildline Autosal 8400-B. Vertical profiles of salinity and temperature were assessed hourly within the channel during a 12-hour period on 1 July. Vertical profiles of temperature, salinity and dissolved O_2 concentrations within the pond were also carried out on four separate days during the study.

Groundwater samples were collected using a drive-point piezometer. Water was pumped using a peristaltic pump at flow rates of ~200 mL min^{-1}, and S, T and dissolved oxygen values were measured using a YSI 600XLM Sonde and recorded. Radon samples were collected, unfiltered, in 250 mL glass bottles by overflowing with three times the bottle volume. Radium samples were collected by pumping 5–10 L of water through MnO_2-coated fibers (Moore, 1976). Groundwater nutrient samples, as well as surface-water nutrient samples, were collected in a syringe and filtered through a 0.45 μm filter into a 15 mL bottle. All nutrient samples were kept on ice after collection and were frozen within 10 h. All apparatus for collection, filtration and storage of nutrient samples were acid-washed prior to use.

Radon analyses of groundwater samples were carried out using a RAD7 radon detector equipped with a sample sparging device that attached directly to the sample bottles. These measurements were carried out typically within a few hours, but no later than two days, after collection. All activities were decay-corrected to the sampling date. Radium analyses were carried out using a well-type gamma detector.

Nutrient samples were analyzed for nitrate and ammonium ion using a Lachat QuickChem 8000FIA autoanalyzer. Samples for total dissolved nitrogen (TDN) were digested prior to analysis using the modified persulfate digestion of D'Elia et al. (1977). Dissolved organic nitrogen (DON) was determined from the difference between TDN and dissolved inorganic nitrogen (DIN=nitrate+ammonium ion).

Seepage meters used in this work were based on the traditional Lee-type seepage meter, (Lee, 1977) made from the top ~30 cm of a 55-gallon drum fitted with outflow and vent ports. Measurements were carried out using methods similar to those described by Shaw and Prepas (1989). Due to the limited number of seepage meters, no measurements were carried out in locations where water depth at low tide was greater than 1 m.

3. Results

The tidal range varied from 0.7 to 1.5 m during the course of the study (Figs. 2a–2c), with spring tides occurring one day after completion of the study (2 July). Wind speeds were low, typically less than 6 m s^{-1} (U_{10}=wind speed at 10 m height) during the day, decreasing to extremely low values at night (Fig. 2d).

Salt Pond was weakly stratified during the study, with the pycnocline depth of roughly one meter and surface water salinities ranging from 30.4 to 30.8, typically 0.1–0.5 psu less saline than the deep waters of the pond. Surface water temperatures ranged from 22 to 24°C, while bottom-water temperature was close to 19°C. The bottom water remained oxic throughout the study, with dissolved oxygen concentrations close to 4 mg L^{-1} (50% of saturation).

Radon activities in the channel reached maximal values of 250–300 Bq m^{-3} after low tide, decreased rapidly to minimum values of ~80 Bq m^{-3} near high tide, and reached intermediate values of 100–150 Bq m^{-3} during the falling tide (Fig. 2a). Salinity values in the channel consistently reached minimum values of 29.5–29.8, on average, 53±10 min after low tide. Salinity increased rapidly with rising tide to maxima of ~30.8–31.1 at high tide, and decreased to values of ~30.5 during the falling tide (Fig. 2b). It is worth noting that the radon maxima occurred ~19±7 min after the salinity minima due to a combination of time required for equilibration of the radon signal in water with the radon in air and time required for ingrowth of ^{218}Po ($t_{1/2}$≈3 min), the radon decay product that is actually measured (see Burnett et al., 2001). For the purpose of consistency with salinity and other data we shifted the time of each radon measurement throughout this work to a value 20 min earlier than measured to correct for this delay. Changes in channel temperatures were less consistent than changes in salinity and radon values. Typically, water inflowing from Nauset Marsh was colder than outflowing water, but values were heavily influenced by daytime heating and nighttime cooling.

Temperature and salinity data both suggest the channel was well-mixed vertically. Spanning the four-day study, water temperatures measured ~23 cm from the bottom of the channel were indistinguishable from the temperature measurements carried out on pumped surface water, with the exception of the intervals when the pump stopped due to clogging. In addition, hourly vertical profiles of temperature and salinity spanning a twelve-hour period revealed no significant stratification, also suggesting the channel was well-mixed.

4. Discussion

The goal of this work from the outset was to quantify groundwater discharge to the pond, using radon and salinity measurements within the channel to construct whole-pond mass balances of radon and salinity and thereby quantify groundwater fluxes to the pond. However, the salinity and radon data from within the channel revealed some unanticipated complexities. Some minutes after low tide, there is consistently a minimum in salinity and a maximum in radon activity. Simultaneous reductions in salinity and increases in radon activities strongly suggest inflow of low-salinity groundwater. There are three pieces of evidence that suggest this event is caused by groundwater discharge directly to the channel, rather than to the pond. First, seepage directly into the channel was observed at more than one location during low tide. Second, the salinity minimum, measured within the channel near Salt Pond, occurred 53±10 min after the low tide, when the tide had risen 9±3 cm. This suggests that the low-salinity, high-radon water was accumulating throughout much of the channel at low tide and was displaced only when the incoming tide carried high-salinity, low-radon water into Salt Pond from Nauset Marsh. Third, during a later sampling period (4–6 August), similar salinity minima were observed just after low tide at both the northern and southern ends of the channel, indicating the phenomenon was not limited to the northern end of the channel where sampling occurred in late June/early July. Together, these data strongly suggest groundwater discharge to the channel at low tide.

4.1. SGD estimated from radon

As a starting point for constraining the groundwater flux to the Salt Pond system from radon measurements, we turn to recent work by Burnett (Burnett and Dulaiova, 2003). Burnett has demonstrated how the velocity of groundwater flow through coastal sediments, integrated over a broad area, can be estimated from the flux of radon to the water column and from the radon activity in the groundwater. In brief,

$$\text{groundwater velocity } (\mathrm{m\,d^{-1}}) = \frac{\text{radon flux } (\mathrm{Bq\,m^{-2}\,d^{-1}})}{[\mathrm{Rn}]_{\mathrm{gw}}(\mathrm{Bq\,m^{-3}})}.$$

Translating this groundwater velocity to a flow rate ($\mathrm{m^3\,d^{-1}}$) requires making assumptions about the area over which this flux occurs, and corrections for mixing and gas exchange.

In this work, we take a slightly different approach to quantifying groundwater flow from radon activities. We take advantage of the narrow channel connecting Salt Pond to Nauset Marsh (Fig. 1) to construct a budget of radon and salinity within the pond. This approach recognizes that we can more reliably measure the flux of radon and salinity to and from the pond through the channel, than the influx of radon, or low-salinity water to the pond via groundwater, which is diffuse and spread out over a large area. Because of the short residence time of the water in the pond (~1.5 d as defined by pond volume/daily outflow), the outflow of radon from the pond is equal to the radon inflow from groundwater, over a timescale of several days, once corrections are made for losses due to gas exchange, decay and inflow from Nauset Marsh (Fig. 3). We can thus infer the groundwater flux to the pond from the measured outflow of radon from the channel.

The flow of water in and out of Salt Pond was assumed to be driven solely by tidal fluctuations and was estimated by multiplying the measured tidal height variations by the surface area of the pond, with the pond surface area changing based on the bathymetry presented in Anderson and Stolzenbach (1985). This approach ignores the effects of wind, which is known to affect circulation in shallow estuaries. However, the presence of a narrow constriction at the mouth, such as exists in Salt Pond (Fig. 1) greatly diminishes any impact of the wind on water flow in and out of an estuary (Geyer et al., 1997). Furthermore, wind speeds were low during the study (Fig. 2d).

Conducting a mass balance for salt based on the measured water inflows and outflows through the channel is fairly straightforward, although the groundwater inputs to the channel complicate the picture. Carrying out a mass balance for radon is more involved, however, due to the additional corrections required for gas exchange and radioactive decay. In order to quantify groundwater discharge to the pond while factoring in these complexities, we use a box model.

4.2. Two box model of Salt Pond

For the purposes of the model, the Salt Pond system is divided into two boxes. Salt Pond itself is treated as one box, while the channel connecting Salt Pond to Nauset Marsh is represented by the second box (Fig. 3). Nauset Marsh is not actually modeled; rather, it is treated as an infinite reservoir of high-salinity, low-radon water adjacent to the channel connecting the marsh to Salt Pond. The dimensions of the boxes are determined in part by the bathymetric map presented in Anderson and Stolzenbach (1985) and partly from aerial photos. Salt Pond surface area, at low tide, is assumed to be equal to 82 200 m^2 (Anderson and Stolzenbach, 1985). The total volume of the pond is assumed to be 280 000 m^3 at low tide. The channel, at low tide, is 30 m across, 350 m long and 0.6 m deep at the thalweg (low point). The volume of the channel is allowed to vary with tidal height according to bathymetry measurements and is roughly five times greater at high tide than at low tide.

This treatment ignores the weak stratification observed within Salt Pond. While this is certainly an oversimplification, we felt that this was a more defensible modeling strategy than to divide Salt Pond into a shallow and deep box and try to model, within the limitations of a simple box model, the complex processes of mixing and entrainment that exchange water between the surficial and deep waters. However, we did create just such a three-box model that could reproduce the observed weak stratification. The discharge estimates from this three-box model were only 15% lower than those we will present for the two-box model, a difference that is within the uncertainty estimate for this approach.

We can summarize the key model assumptions as follows. Pond inflows and outflows are driven entirely by tidal height fluctuations (which were measured). The radon content and salinity of Nauset Marsh waters are assumed constant ([Rn]=80 Bq m^{-3}, S=30.9). The channel is assumed well-mixed (see results section). For the sake of initial simplicity, groundwater salinity is assumed to equal zero (this assumption will be assessed later). The average radon activity measured in fresh groundwater (9400±4000 Bq m^{-3}) is assumed representative of groundwater inflow (n=~20). It is worth noting that there was less uncertainty in the fresh groundwater radon activities collected within 1.5 m of the sediment surface (7200±800 Bq m^{-3}, n=7), but for now we will assume the average value of all fresh groundwater estimates. Radon diffusion from sediments is assumed to be negligible, which will be demonstrated later. Gas exchange in the channel is ignored (because the radon content of water inflowing from Nauset Marsh is assumed constant). We ignore precipitation and evaporation, which are negligible during the study. We also ignore changes in radon and salt storage within Salt Pond during the four-day study. The short residence time of the pond, together with the very consistent salinity and radon data measured within the channel, suggest that no major storage changes occurred.

Radon losses due to radioactive decay and to gas exchange (within the pond) require additional explanation. Loss due to decay is treated as a first-order process based on the radon half-life of 3.82 days, assuming there is no supporting ^{226}Ra. This assumption is fairly well justified, as Rn activities range from 80–300 Bq m^{-3} (Fig. 2), while activities of the parent isotope, ^{226}Ra, range from 1–2 Bq m^{-3}. Radon loss by gas exchange was assumed to be due solely to wind and was estimated using the wind speed and Schmidt number dependence of Turner et al. (1996; also see Appendix). This is an oversimplification, as gas exchange has been observed to vary with current speed in estuaries (Zappa et al., 2003). However, as Salt Pond has no river flowing into it, and it is the most landward portion of the Nauset Marsh system (Fig. 1), current speeds are likely to be much lower than in many estuaries. Furthermore, radon measurements carried out on Salt Pond surface waters reveal a strong inverse relationship between radon activities and windspeed (Fig. 4). We cannot accurately model these data because tidal height measurements were not carried out simultaneously. However, the strong inverse relationship implies an important impact of wind speed on gas transfer velocities.

In the interest of clarity and brevity the model equations are presented in the Appendix. Model parameter values are based primarily on measurements and observations. The true unknowns include only the magnitude of the groundwater flows to the pond and to the channel and the dependence of these flows on tidal height.

In order to convey a sense of the controls on the channel radon and salinity data we will systematically discuss a series of model sensitivity tests. For all of the model runs we assume groundwater discharge is characterized by a salinity of zero (we will evaluate this assumption later). For all current model runs we will also assume discharge occurs only when the tidal height is within ten cm of low tide. Later, we will test the sensitivity to this assumption.

When groundwater is discharged only to the channel, but not to the pond, the model does a reasonable job of reproducing the timing and magnitude of the radon maxima and salinity minima (Figs. 5a and 5b; ignoring the magnitude of the flow for the moment). However, this groundwater input to the channel cannot be very important to the overall radon and salinity budgets for the pond as a whole. Although this discharge to the channel is carried into the pond with the incoming tide during the model simulation, the modeled radon values during the outgoing tides are lower than the observed radon values, while the modeled salinity values are higher than observed (Figs. 5a and 5b). This suggests there must be an additional source of groundwater discharging to the pond itself.

When groundwater is discharged only to the pond, but not to the channel (also ignoring the flow rate for the moment), the model does a reasonable job of simulating the radon activity and the salinity of the outflowing water during falling tides (Figs. 5c and 5d). However, the model does not reproduce the radon maxima and salinity minima observed just after low tide. Groundwater discharge to both the channel and to the pond are required to reproduce the full dataset adequately.

Having demonstrated how groundwater inputs to the channel and to the pond manifest themselves in the channel data, we combine these inputs and use the model to test the sensitivity to flow to the channel and to the pond. We first hold groundwater flow to the pond constant (at a value of 2200 $m^3 d^{-1}$) but allow groundwater discharge to the channel to vary. When channel groundwater inflow is 160 $m^3 d^{-1}$ the radon maxima at low tide are clearly too low while the salinity minima are clearly too high, suggesting this is an underestimate of the true flow (Figs. 6a and 6b). By contrast, modeled flow of 800 $m^3 d^{-1}$ to the channel is clearly an overestimate. Modeled flow of 300 $m^3 d^{-1}$ yields a reasonable (although not perfect) fit to the data.

If we maintain the modeled flow to the channel at a flow rate of 300 $m^3 d^{-1}$ we can test the sensitivity to flow to the pond. When model groundwater discharge to the pond is set to 1100 $m^3 d^{-1}$, the radon values during the falling tides are clearly too low, while the salinity values are too high (Figs. 6c and 6d). This modeled groundwater input to the pond is clearly an underestimate. By contrast, groundwater inflow of 4200 $m^3 d^{-1}$ is clearly too high (Figs. 6c and 6d). It is worth noting that this high figure is the long-term average discharge predicted by a well-developed hydrologic model (Colman and Masterson, 2004), although other parameterizations of that same model predict 3200 $m^3 d^{-1}$. Groundwater inflow to the pond of 2200 $m^3 d^{-1}$ yields a reasonable fit to both the radon and salinity data (Figs. 6c and 6d).

Model runs to this point have all assumed groundwater discharge only occurs when the tidal height is within ten cm of low tide. We now use the model to test this assumption. When discharge to the channel occurs only within ten centimeters of low tide, the low-tide radon maxima and salinity minima are brief events that match the data reasonably well (Figs. 7a and b). However, when discharge to the channel is allowed to occur within thirty centimeters of low tide, the radon maxima and salinity minima are much broader, longer-lasting features that deviate substantially from the data. These results may suggest that discharge to the channel is limited to the times close to low tide, although it is also possible that this phenomenon is due to processes that are not well represented by this simple model.

Similarly, we should be able to use the model to assess the sensitivity of the data to the timing of groundwater discharge to the pond itself. In this case there is very little difference between the model runs with discharge occurring within 10 cm of low tide, within 50 cm of low tide, and at all tidal heights (Figs. 7c and 7d). The primary reason the model does not suggest a strong dependence of discharge on tidal height in the pond itself is because the pond volume is large relative to the length of the shoreline, where groundwater discharge is focused (see seepage meter data below). By contrast, the channel volume is much smaller, despite a similar length of shoreline, which serves to magnify the temporal variability in the radon and salinity data with time-varying groundwater inputs. Keep in mind that this model is evaluating the average pond radon activity. As has been observed in a number of studies, near-shore radon activities often do vary with tidal height (Burnett and Dulaiova, 2003; Lambert and Burnett, 2003; Abraham et al., 2003) because mixing is not instantaneous, as is assumed in this box model.

Summarizing, modeled inflows of roughly 300 $m^3 d^{-1}$ to the channel, and 2200 $m^3 d^{-1}$ to the pond, yield a reasonable fit to all of the salinity and radon data from the channel (Figs. 8a and 8b), indicating a net inflow to the Salt Pond system of roughly 2500 $m^3 d^{-1}$. This translates to an average flow velocity to the pond of 3 cm d^{-1}. Due largely to the uncertainty in the radon activity in groundwater, we assign an uncertainty of roughly 50% to these estimates. While the model fits are fairly good for the radon data, they are not always so good for the salinity simulations. The simple reason for this is that the channel salinity does not behave in a consistent manner during falling tides. Sometimes salinity increases during the falling tide (Fig. 8b; day ∼1.7), sometimes it remains constant (day ∼1.1, ∼2.7) and sometimes it decreases (day ∼2.1, ∼3.2, ∼3.8). Decreases in salinity could be explained by increased input of groundwater at lower tides. However, if this were the correct explanation one would expect increases in radon activity at the same time. While gaps in the radon data limit our ability to assess this possibility fully, one interval of gradually lowering salinity during falling tide near day 3.8 shows no such corresponding increase in radon activity. Thus, the reasons for the inconsistent trends in salinity during the falling tides remain unclear.

It is worth noting that the only rainfall that occurred during the study period was on 29 June (between integer days 1 and 2 on Fig. 2), when 2.5 mm of rain fell. We note that the measured salinity values within the channel on this day were among the lowest values measured during the study. However, because we do not know the timing of any freshwater inputs from runoff or extremely rapid groundwater inputs, we elected not to factor this rainfall event into the model.

With these best-fit model data (Figs. 8a and 8b) the relative sizes of the loss terms can be evaluated as a tool for understanding the largest uncertainties (Fig. 8c). By far the largest loss term in the radon budget is outflow from the pond. Losses due to gas exchange and decay are small, due to low wind speeds (Fig. 2d) and the fairly short residence time of the pond (∼1.5 days). Therefore, even if the gas exchange loss estimate is low (for example if gas exchange due to flow-induced turbulence were significant) the radon-derived groundwater flow estimates would not be significantly different. These loss terms must be comparable in magnitude to the source terms for radon, in order for mass balance to be conserved. It is thus worth noting that typical diffusive inputs of radon in geologically similar areas are on the order of 1–10 $\mathrm{Bq\,m^{-2}\,d^{-1}}$ (Hussain et al, 1999; Schwartz et al., 2003), much lower than the overall inputs (Fig. 8c). We thus conclude that diffusive inputs from sediments are negligible.

4.3. SGD estimated by seepage meters

Seepage meter data from Salt Pond offer an additional constraint on the dependence of groundwater discharge on tidal height, and on distance from shore (or water depth in the pond). It is important to keep in mind that seepage meter flow estimates are, at best, representative of the small area covered by the seepage meter (<1 m^2). Furthermore, seepage meters have been criticized as prone to artifacts (Shaw and Prepas, 1989; Shinn et al., 2002). However, recent intercomparisons between seepage meters and other means of estimating SGD suggests seepage meters can give reasonable SGD estimates when used properly (Corbett and Cable, 2003; Lambert and Burnett, 2003; Taniguchi et al., 2003).

In this work, a total of ten seepage meters were deployed on two successive days. On the first day, seepage meters were deployed in three transects parallel to the shore, at an average water depth of 0.5 m below low tide. On the second day, the seepage meters were deployed in three transects perpendicular to shore, at water depths ranging from ~0.2 to ~0.7 m below low tide. One important feature of the data that stands out is that there was considerable variability among the meters (Figs. 9a and 9b), particularly when the seepage meters were deployed in three transects perpendicular to the shore (Fig. 9b). Another feature of note is that there is always discharge into the pond, even when the tidal height is 0.5 m above low tide. Furthermore, for many of the seepage meters, but not all, discharge increases slightly at low tide (Figs. 9a and 9b). One important trend from the transects perpendicular to shore helps to explain some of the variability. In two of the three transects, greater discharge (≥20 cm d^{-1}) occurred at shallow sites (closer to shore) than at deeper sites (farther from shore) (Fig. 9c). Flow was greatly diminished (to <10 cm d^{-1}) at water depths 0.5–0.7 m below low tide that were farther from shore (Fig. 9c). It is difficult to extrapolate to the whole pond based on these limited data, given the large variability and the fact that the ten seepage meters spanned only ~0.003% of the pond surface. Nonetheless, taken at face value, these data suggest that discharge occurred in a thin band close to shore. Indeed, shallow discharge is also implied by electrical resistivity data (Bratton et al., in preparation) which indicate fresh water in the subsurface only in the shallow sediments of a transect perpendicular to shore.

4.4. Comparisons of SGD techniques

The seepage meter discharge estimates, while limited in areal and temporal extent, can be compared to the flow estimate based on radon and salinity. Only 20% of the surface of the pond would experience discharge if we assume that discharge is limited to sediments shallower than one meter water depth (at low tide), as suggested by the seepage meter results. If we multiply the average seepage-meter-inferred groundwater velocity of 16 cm d^{-1} (from the transects perpendicular to shore), by the area of discharge (16 000 m^2) through these shallow sediments, we obtain a flow rate of 2900 m^3 d^{-1}. This figure is very similar to the estimate based on modeling of radon and salinity data of 2500±1250 m^3 d^{-1}. It is important to bear in mind, however, that the seepage meters were deployed a week after the channel experiment was conducted.

The measured discharge rate of 2500±1250 m^3 d^{-1} based on the radon and salinity data translates to a groundwater velocity of 3 cm d^{-1} averaged over the entire pond, which is similar to other estimates of groundwater velocity from geologically similar settings. For example, the groundwater discharge velocity in nearby Waquoit Bay was estimated to be ~8 cm d^{-1} (Abraham et al., 2003). Furthermore, the discharge rate agrees fairly well with an estimate of 3200–4500 m^3 d^{-1} based on hydrologic flow modeling to Salt Pond (Masterson, 2004; Colman and Masterson, 2004), given the uncertainties in each approach. Despite the agreement, additional work is necessary to evaluate both how representative this discharge rate is of the long-term mean as well as the rate of discharge seaward of Salt Pond. In this light, it is worth noting that a thirty-year record of monthly water-table elevation data is available from a well in nearby Eastham, MA. The water table elevations from the ~8 month interval prior to our study period are intermediate values compared to the full thirty-year record (data not shown). This may suggest that discharge during the period of our study was representative of the long-term mean used in the hydrologic model.

One might expect SGD estimates based on radon to exceed estimates of fresh groundwater discharge, since the radon flux to surface waters could include both a fresh and a saline component. In this work, however, there is no need to invoke any contribution to radon from saline groundwater because the general features of the radon and salinity trends are reproduced by the box model assuming only inputs from fresh groundwater. There are at least two points worth discussing in this light. The first observation is that, as mentioned earlier, organic-rich, impermeable sediments at the center of the pond may hinder the movement of any water through sediments underlying a large portion of the pond. A consistent hydraulic gradient may thus drive flow of fresh groundwater through shallow sediments at all times and minimize recirculation of seawater via tidal pumping. Such a phenomenon would help explain why seepage meter data suggest discharge to the pond at all tidal heights, and why fresh groundwater was observed to depths of a few meters in a number of the piezometers sampled that were underlying saline surface water (data not shown).

The second observation is that a plot of radon activities versus salinity in the groundwater (Fig. 10) reveals higher radon content in the fresh groundwater (average=9400 Bq m^{-3}) than in the saline groundwater (average=4000 Bq m^{-3}). This may suggest that the radon content of saline groundwater is lower than in fresh groundwater at this site in general, or it may suggest that radon has recently been removed from the saline groundwaters due to recent advection. In either case there would be a smaller flux of ^{222}Rn from saline groundwater than from fresh. It is worth noting that recent work in nearby Waquoit Bay (Abraham et al., 2003) revealed higher, rather than lower, radon activities at high salinity. Further work is clearly needed to better understand the influences of salinity, tides and geochemical processes on radon (and its parent isotope ^{226}Ra) in submarine groundwaters.

4.5. Groundwater-derived nutrient discharge

Most domestic wastewater on Cape Cod is treated using septic systems, which has led to significant nutrient discharges to groundwater and consequently to coastal eutrophication. This issue has drawn increasing attention from researchers, as well as communities, in recent years (Valiela et al., 1990; Giblin and Gaines, 1990; Portnoy et al., 1998; Nowicki et al., 1999; Charette et al., 2001; Colman and Masterson, 2004). This significant regional concern motivated this attempt to quantify groundwater-derived nutrient discharge to Salt Pond, focusing on nitrogen, the limiting nutrient in most estuaries. Total dissolved nitrogen (TDN) concentrations in Salt Pond groundwater averaged 93 μmol kg^{-1} (n=57) and were composed of 61% NO_3^-, 14% NH_4^+ and 32% dissolved organic nitrogen (DON), on average. Multiplying these concentrations by the average groundwater flux inferred from modeling the radon and salinity data yields an average influx to Salt Pond of 2.6 mmol m^{-2} d^{-1} of TDN from groundwater. This value is lower than the figure of 11.9 mmol m^{2} d^{-1} predicted by Colman and Masterson (2004), partly because of the lower-than-predicted groundwater fluxes and partly because the nutrient concentrations measured in groundwater were lower than assumed in the model. The average flux of nitrogen from groundwater to nearby Town Cove was estimated by Giblin and Gaines (1990) to be 1.8 mmol m^{-2} yr^{-1}, a figure that is very close to our estimate. A different study measured nitrate fluxes in seepage meters that were more than an order of magnitude higher (Portnoy et al., 1998). However, the seepage meter-derived estimates are from discharge sites of limited but unknown areal extent and therefore overestimate the average influx to Town Cove. For comparison, it is worth contrasting these nutrient discharge estimates from Cape Cod with the average TDN discharge rate of 2.6 mmol m^{-2} d^{-1} to Chesapeake Bay (Chesapeake Bay Program, 1999; Charette and Buesseler, 2004), a site well known to be heavily impacted by anthropogenic nutrient inputs. The Salt Pond nutrient flux estimates from this work merely add to the growing body of knowledge illustrating that groundwater-derived nutrient loads are a significant environmental problem on Cape Cod.

5. Conclusions

This work adds to the growing evidence that continuous radon measurements offer a powerful means to constrain submarine groundwater discharge to coastal waters. Radon data from the channel connecting Salt Pond to Nauset Marsh can be modeled assuming inputs of fresh groundwater to both the channel and to the pond, with no need to invoke a saline component. The absence of a saline component in the radon flux may be due to removal of radon from saline groundwater by recent advection of seawater or it may be due to the presence of impermeable sediments in the center of the pond that limit the opportunities for recirculation of seawater through much of the sediment.

Box modeling of radon and salinity data implies groundwater discharge of 300±150 $m^3 d^{-1}$ to the channel and 2200±1100 $m^3 d^{-1}$ to the pond for a total discharge of 2500±1250 $m^3 d^{-1}$ to the Salt Pond system. This translates to an average groundwater flow velocity through the pond sediments of ∼3 cm d^{-1}. The discharge estimate is similar to that of 3200–4500 $m^3 d^{-1}$ predicted by a hydrologic model (Colman and Masterson, 2004; Masterson, 2004). Further research is needed to determine how representative this discharge estimate is of the long-term mean, and to determine the rate of discharge seaward of Salt Pond. Data also suggest a TDN flux from groundwater to Salt Pond of ∼2.6 mmol $m^{-2} d^{-1}$, a figure comparable to the average TDN flux from all sources to eutrophic Chesapeake Bay.

Appendix: Salt Pond Box model equations

For the purpose of the box model the Salt Pond "system" is divided into a box representing the pond and a box representing the inlet channel (Fig. 4). Nauset Marsh is treated as an infinite source of water with fixed salinity and radon activity. The equations describing sources and sinks of salt and radon in each of the boxes are presented below. For bookkeeping purposes, each location modeled is given a number according to: gw=1, pond=2, channel=3, Nauset=4.

$$\frac{dS_2}{dt}=(Q_{32}(S_3-S_2) + Q_{23}(S_2-S_3) + Q_{12}(S_1-S_2))/V_2 \tag{A1}$$

$$\frac{dRn_2}{dt}=(Q_{32}(Rn_3-Rn_2)+Q_{23}(Rn_2-Rn_3)+Q_{12}(Rn_1-Rn_2)-\lambda Rn_2V_2-kA_2Rn_2)/V_2 \quad \text{(A2)}$$

$$\frac{dS_3}{dt}=(Q_{43}(S_4-S_3)+Q_{23}(S_2-S_3)+Q_{13}(S_1-S_3))/V_3 \quad \text{(A3)}$$

$$\frac{dRn_3}{dt}=(Q_{43}(Rn_4-Rn_3)+Q_{23}(Rn_2-Rn_3)+Q_{13}(Rn_1-Rn_3)-\lambda Rn_3V_3)/V_3. \quad \text{(A4)}$$

S_i=salinity in box i
Rn_i=radon activity in box i ($Bq\,m^{-3}$)
Q_{ij} = water flux from box i to box j ($m^3\,d^{-1}$)
λ=^{222}Rn decay constant=0.181 d^{-1}
V_i=volume of box i (m^3)
k=gas transfer velocity ($m\,d^{-1}$).

We use the dependence on wind speed presented by Turner et al. (1996), where $k_{600}=0.45\mu^{1.6}$ $(Sc/600)^{-a}$, where μ is wind velocity ($m\,s^{-1}$), Sc is the Schmidt number for radon at the desired water temperature, and a is a variable exponent that equals 0.6667 for $\mu\leq3.6\,m\,s^{-1}$ and equals 0.5 when $\mu>3.6\,m\,s^{-1}$. The number 600 is the Schmidt number for CO_2 at 20°C, a common reference point.

A_2=surface area of Salt Pond = 82,200 m^2 at low tide
A_3=surface area of channel (m^2).

It is worth noting that we ignore the effects of evaporation, because its impact is minor. For simplicity we also ignore the impact of precipitation. A total of 2.5 mm of rain fell on 29 June. This may have contributed to low salinity values observed during that day. However, we do not know the timing and magnitude of inputs from runoff and shallow groundwater flow, hence we ignore the effect of rainfall.

Acknowledgements. For assorted logistical support we thank J. Portnoy and N. Finley of the National Park Service and H. Lind and his crew from the Eastham, MA Dept. of Natural Resources. We thank the summer fellowship program of WHOI for support of L. Ryckman and the MBL summer fellowship program for support of K. Halloran. Thanks to A. Rago (WHOI) and to S. Baldwin and J. Palardy for help in the field, and to J. Warner for discussions of estuarine mixing processes and modeling. Financial support was provided by the U.S. Geological Survey and by National Science Foundation grant #OCE-0346933 to MAC.

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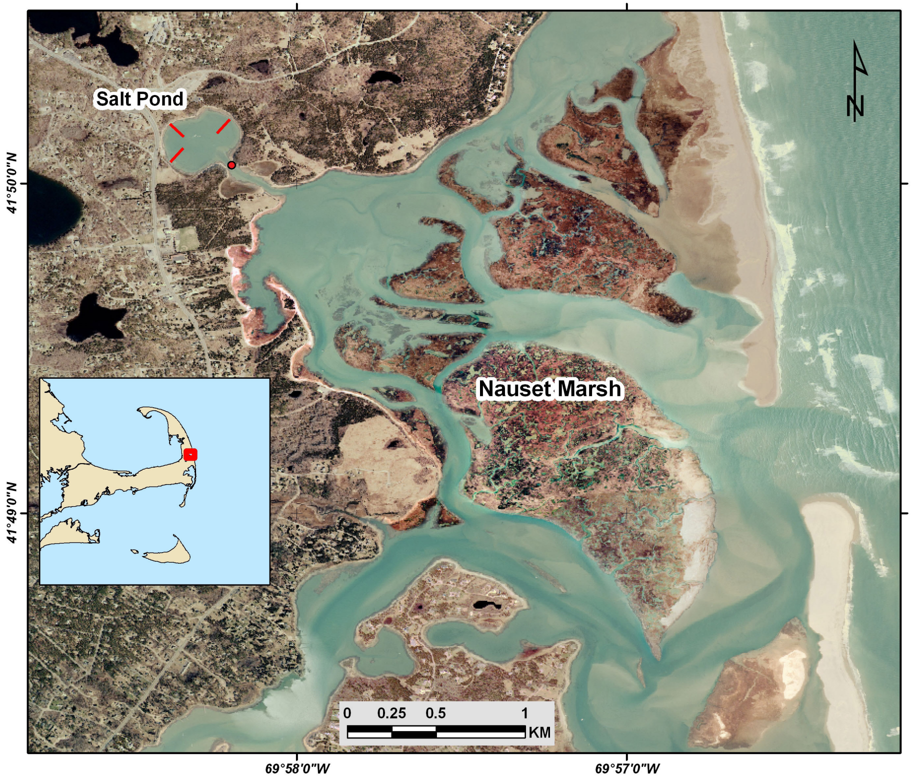

Fig. 1. Aerial photo of Salt Pond and Nauset Marsh system (Cape Cod location on inset), showing seepage meter locations as red lines perpendicular to shore. Transects were less than 10 m long but are exaggerated in the figure for the sake of clarity. The sampling raft location is also shown as a red dot at the NW end of the channel (69.97063˚N, 41.83432˚W).

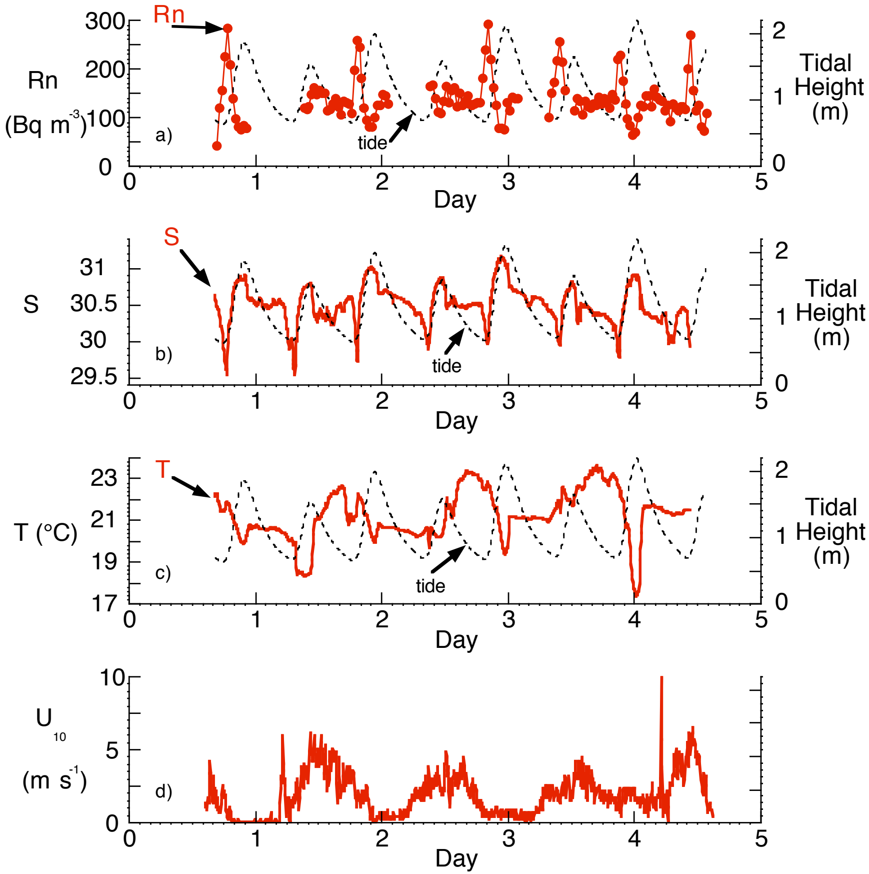

Fig. 2. **(a)** Radon, **(b)** salinity, **(c)** temperature, and **(d)** wind speed data from 28 June–2 July 2004, from the channel between Salt Pond and Nauset Marsh (location shown in Fig. 1). The integer day values correspond to midnight. Channel water depth at the measurement point (a measure of tidal height) is also shown on (a)–(c) as a dashed line.

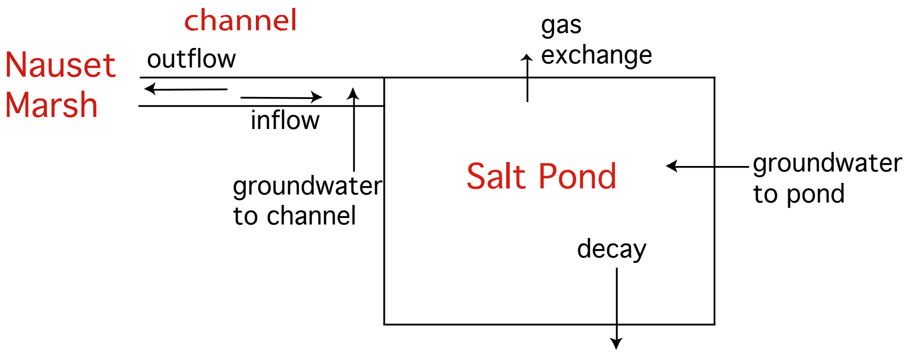

Fig. 3. Schematic indicating the major sources and sinks of radon in the box model of the Salt Pond system.

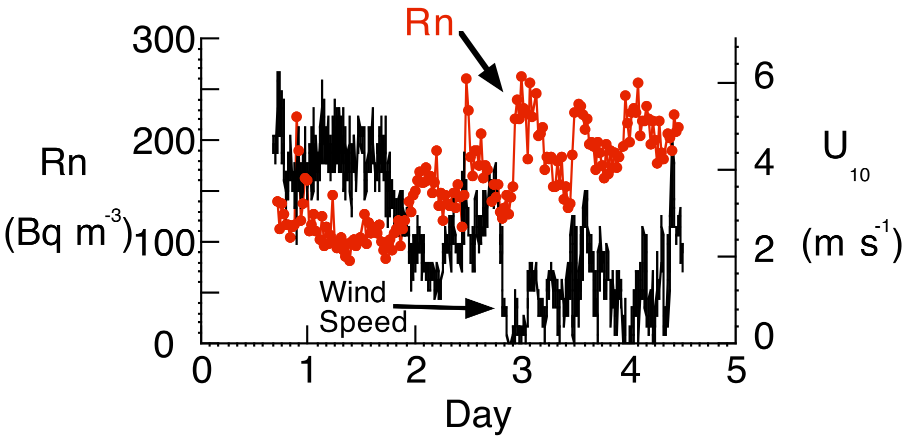

Fig. 4. Surface-water radon data, and wind speeds (10 m height) from the center of the pond, from the week of 14–18 June.

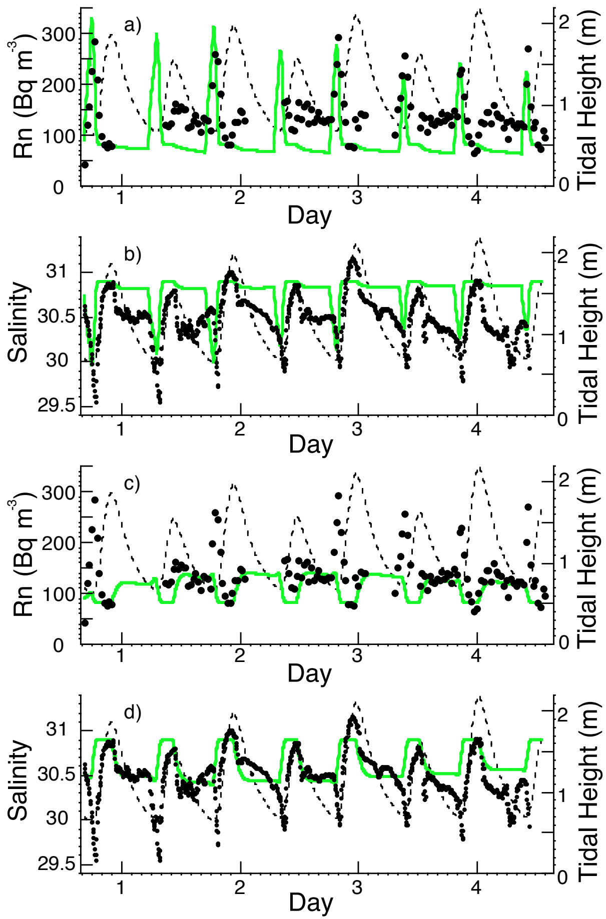

Fig. 5. Box model simulation of channel radon and salinity data assuming discharge to the channel only **(a)**, **(b)** and the pond only **(c)**, **(d)**. Model simulations are portrayed in green, data are presented as solid circles and tidal height is presented as a dashed line.

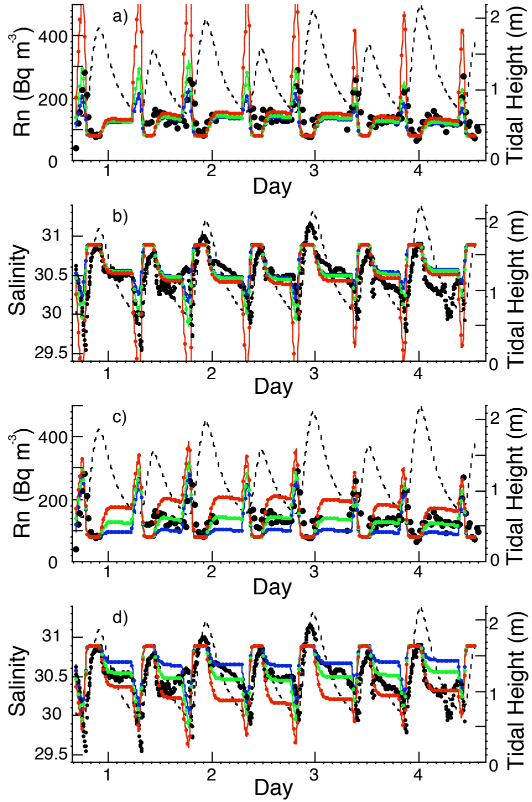

Fig. 6. Box model sensitivity test of channel radon and salinity data assuming: **(a), (b)** Groundwater discharge to the pond is held constant at a value of 2200 $m^3 d^{-1}$ while discharge to the channel is allowed to vary from 160 $m^3 d^{-1}$ (blue), 300 $m^3 d^{-1}$ (green) and 800 $m^3 d^{-1}$ (red); **(c), (d)** Groundwater discharge to the channel is held constant at a value of 300 $m^3 d^{-1}$ while discharge to the pond is allowed to vary from 1100 $m^3 d^{-1}$ (blue), 2200 $m^3 d^{-1}$ (green) and 4200 $m^3 d^{-1}$ (red).

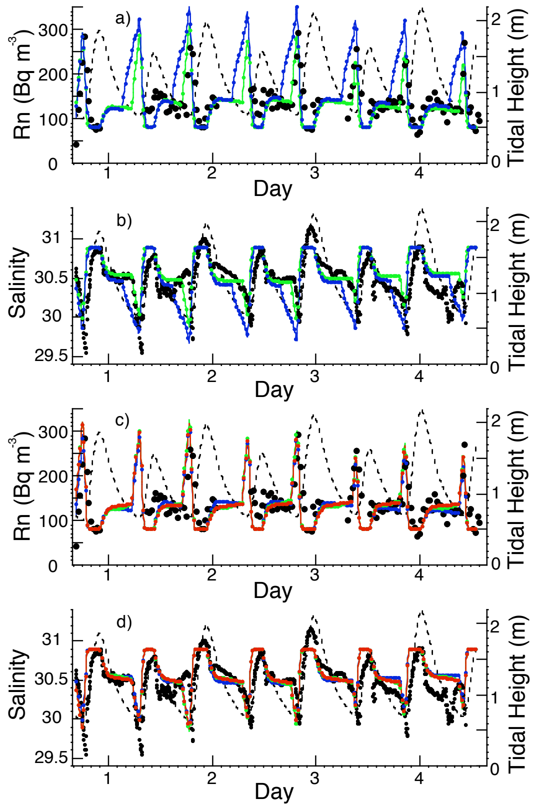

Fig. 7. Assessment of box model sensitivity to tidal height dependence of discharge, assuming: **(a), (b)** groundwater is discharged to the pond within 10 cm of low tide and to the channel within 10 cm (blue) and 30 cm (green) of low tide; **(c), (d)** groundwater is discharged to the channel within 10 cm of low tide and to the pond within 10 cm (blue) and 50 cm (green) of low tide, and at all tidal heights (red).

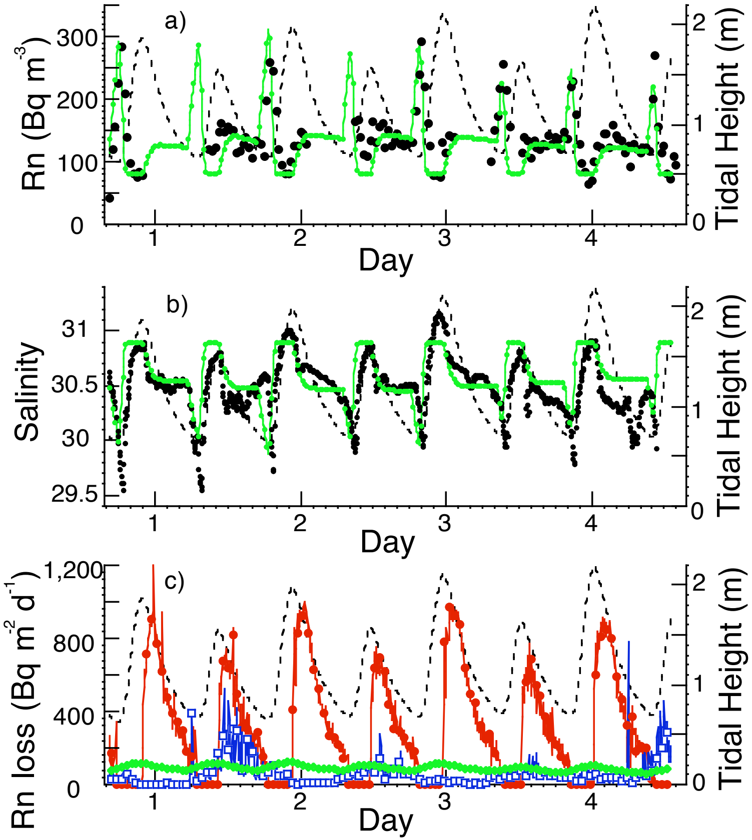

Fig. 8. A "good fit" of the radon and S data **(a), (b)**, assuming groundwater discharge of 300 $m^3 d^{-1}$ to the channel within 10 cm of low tide, discharge of 2200 $m^3 d^{-1}$ to the pond (also within 10 cm of low tide). Also shown are the radon loss terms for this best-fit scenario **(c)**, including loss due to outflow (red), gas exchange (blue), and decay (green), as well as the tidal height (dashed line).

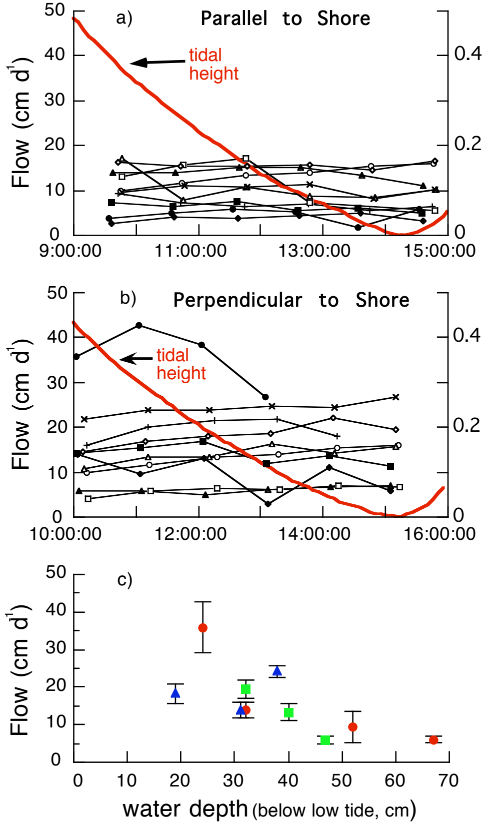

Fig. 9. Groundwater flow velocity estimated using seepage meters aligned in transects parallel to shore **(a)**, and perpendicular to shore **(b)**. Also shown is tidal height (a), (b). The dependence of the average seepage velocity on water depth (below low tide) is also shown from transects perpendicular to shore **(c)**, with each transect noted by a different color symbol.

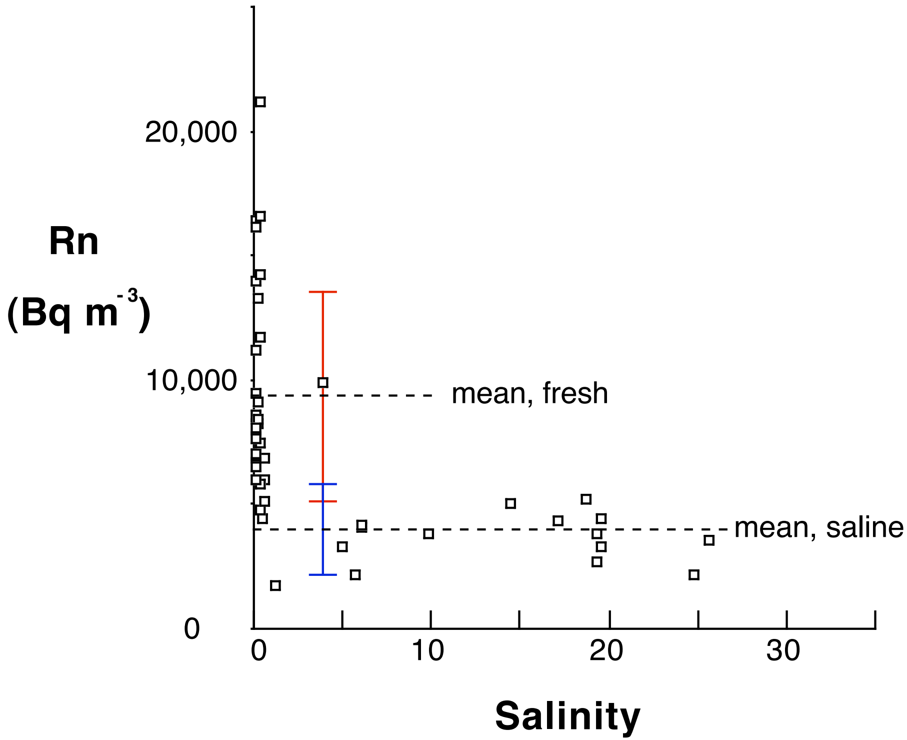

Fig. 10. Groundwater radon activities plotted versus salinity. One-sigma error bars are indicated for both freshwater (S<1) samples (red) and saline (S>1) samples (blue). It is worth noting that the mean for freshwater samples collected within 1.5 m of the sediment surface was 7200±800 Bq m^{-3} (not shown).